

Long-term Contaminant Trends and Patterns in Puget Sound, the Straits of Juan de Fuca, and the Pacific Coast

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Abstract

Existing data on contaminants in dated sediment cores from Puget Sound, and in mussels from the entire U.S. Pacific Coast (65 sites), including Puget Sound (15 sites), were re-examined to evaluate geographical patterns, long-term trends, and signs of effectiveness of contaminant management actions. As previously reported, the dated cores reveal a century-long (1890 -1990) rise and fall of chemical contamination in the Puget Sound. Further examination also reveals possible long-term processes that may have transported materials, such as arsenic, across basins. The Mussel Watch Program data begin where the core ends (late 1980's). Both cores and mussels from the Sound confirm that PCB concentrations have been declining over the past three to four decades. However, there have been no trends for most metals. Compared to mussels from other Pacific coastal sites those within Puget Sound and the Straits contain lower concentrations of several metals (arsenic and cadmium), similar concentrations of other metals, concentrations of PCBs that are similar to other urban areas, but extremely high concentrations of PAH's. To understand and derive benefit from contaminant management actions; and, ultimately, to determine "how clean is clean enough?", these kinds of long-term region-wide contaminant monitoring programs should be maintained, if not enhanced.

Introduction

During the past two decades the National Ocean Service (NOS) of the National Oceanic and Atmospheric Administration (NOAA) conducted several projects in Washington State marine waters as part of the National Status and Trends (NS&T) Program. These include the National Mussel Watch Program (O'Connor 1998), which has monitored contaminants in Washington since 1986, and a Puget Sound Main Basin dated sediment core project, which provided information on contaminant trends dating back to the late 1800's (Lefkovitz and others 1997).

I frequently refer to data from these programs to quickly identify background information during spills of oil and hazardous materials (eg., Mearns and others 1998 and 1999). During the course of this work it became apparent that many people are either not aware of these programs or find it difficult to easily access or interpret the information. I was motivated to help solve this problem by preparing this report.

The purpose of this paper is to bring renewed attention to these chemical contaminant data because collectively they tell a fascinating long-term story about pollution trends in Puget Sound and adjacent coastal areas. I attempt to do this by displaying the existing data in forms that make it a little easier to spot geographic patterns and long-term trends. I prepared graphics so that general patterns (increases, decreases, cycles, etc), if they existed, could be seen visually in the time series plots. It was beyond the scope of the project to conduct a detailed statistical analysis of trends and geographical differences: those interested in statistical confirmation of trends and patterns need to acquire the data and conduct appropriate analyses.

To provide additional perspective on conditions in Puget Sound and the Northwest Straits I also examined contaminant data from several sites along Washington's Pacific Coast. One of these sites, Cape Flattery, may be indicative of contaminant trends in the Olympic Coast National Marine Sanctuary (NMS).

Approach and Methods

Contaminants Considered

Chemical contaminants reviewed for this report include 15 elements (metals), polychlorinated biphenyls

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(PCBs), chlorinated pesticides (DDT, chlordane, dieldrin) and related organic compounds (chlorobenzenes), an organophosphate pesticide (chlorpyrifos), polycyclic aromatic hydrocarbons (PAHs up to 44 compounds), organotin compounds, conventional materials (total organic carbon, nitrogen and phosphorus) and several chemical markers (hopane, terpenes and linear alkyl benzenes).

Mussel Watch

In 1986 NOAA's National Ocean Service (NOS) began monitoring chemical contaminants in mussels and oysters annually from over 200 fixed intertidal locations around the United States. Over the past 15 years the program sampled up to 17 sites in Puget Sound, the Northwest Straits and along the state's Pacific coast (Table 1 and Figure 1). Lauenstein and others (1997) provided detailed descriptions of each site.

Table 1. Code, name, location description, county, latitude, longitude and species sampled for 18 NOAA National Status and Trends, National Mussel Watch Program, Sites in Washington and Oregon. M. = Mytilus.

Site	Location General	Specific	County	Lat N	Long W	Species
<i>COAST:</i>						
CRSJ	Columbia River	South Jetty	Clatsop	46.2333	124.0463	M. californianus
CRNJ	Columbia River	North Jetty	Pacific	46.2778	124.0621	M. californianus
WBNA	Willapa Bay	Nahcotta	Pacific	46.5080	124.0060	M. edulis
GHWJ	Grays Harbor	Westport Jetty	Grays H	46.9125	124.1175	M. californianus
JFCF	Juan de Fuca Str.	Cape Flattery	Clallam	48.3883	124.7213	M. californianus
<i>NORTHWEST STRAITS:</i>						
PSPA	Puget Sound	Port Angeles	Clallam	48.1397	123.4168	M. edulis
PSPT	Puget Sound	Port Townsend	Jefferson	48.1030	122.7650	M. edulis
PSHC	Puget Sound	Hood Canal	Jefferson	47.8317	122.6866	M. edulis
BBSM	Bellingham Bay	Squalicum Jetty	Skagit	48.7542	122.4995	M. edulis
PRPR	Point Roberts	Point Roberts	Skagit	48.9817	123.0216	M. edulis
<i>PUGET SOUND BASIN:</i>						
SSBI	South Puget Snd.	Budd Inlet	Thurston	47.1005	122.9121	M. edulis
CBTP	Commencement	Tahlequah Point	Mason	47.3358	122.5016	M. edulis
PSSS	Puget Sound	South Seattle	King	47.5288	122.3986	M. edulis
EBDH	Elliott Bay	Duwamish Head	King	47.5758	122.4180	M. edulis
EBFR	Elliott Bay	Four-Mile Rock	King	47.6392	122.4123	M. edulis
SIWP	Sinclair Inlet	Waterman Point	Kitsap	47.5508	122.6270	M. edulis
WIPP	Whidbey Island	Possession Point	Island	47.9025	122.3800	M. edulis
PSEH	Puget Sound	Everett Harbor	Snohom.	47.9738	122.2370	M. edulis

Sampling was conducted in the winter, before mussels spawn. From 1986 through 1991 three composites, each containing 30 to 50 mussels were sampled from each site annually. From 1992 to the present, effort was reduced so that only one composite has since been sampled at each site; in 1994 the frequency was reduced to one sampling event every two years. During the course of the program some sites have been dropped and others added.

The NS&T program contractors extracted and analyzed contaminants from composited whole soft tissue of mussels and oysters. Processing and analytical methods are described in detail in four NOAA methods documents (Lauenstein and Cantillo 1993). Details of specific analytes measured are reviewed in several reports such as O'Connor (1998). The program also conducts quality assurance and quality control projects. After receipt, proofing and quality control, chemical data from the program are entered into the NS&T's website (begin with <http://noaa.gov>).

To conduct this project I downloaded all available raw data (1986 through 1998). In some cases (eg., silver) I replaced zero values with either Method Limits of Detection (LODs) or the lowest reported value; otherwise, zero concentrations were used where reported (eg., PAHs). For several contaminant classes

(PCBs, PAHs, DDTs, chlordanes) values from individual analytes were summed to yield “total” values (eg., T PCB, T PAH, etc.). However, not all analytes for a contaminant group were analyzed every year. For example, at the onset of the program only 18 PAHs (mainly parent compounds) were reported. However, by 1993 the Program was reporting up to 44 individual PAH compounds (Mearns and others 1999). Therefore, for long-term (time series) plotting, I used the maximum number of analytes common to all survey years (eg. 18 PAH’s). However, for mapping recent data (1997-98), I used all available analytes.

To observe large-scale (region-wide) geographic patterns, I plotted the most recent available data (1997 through 1998) on maps that included the coastal as well as inland sites. From these data I calculated means, medians, standard deviations and ranges of chemical concentrations and compared these with comparable data from the entire national database for this period. To examine general long-term trends I plotted all individual composite values for all stations on time series graphs.

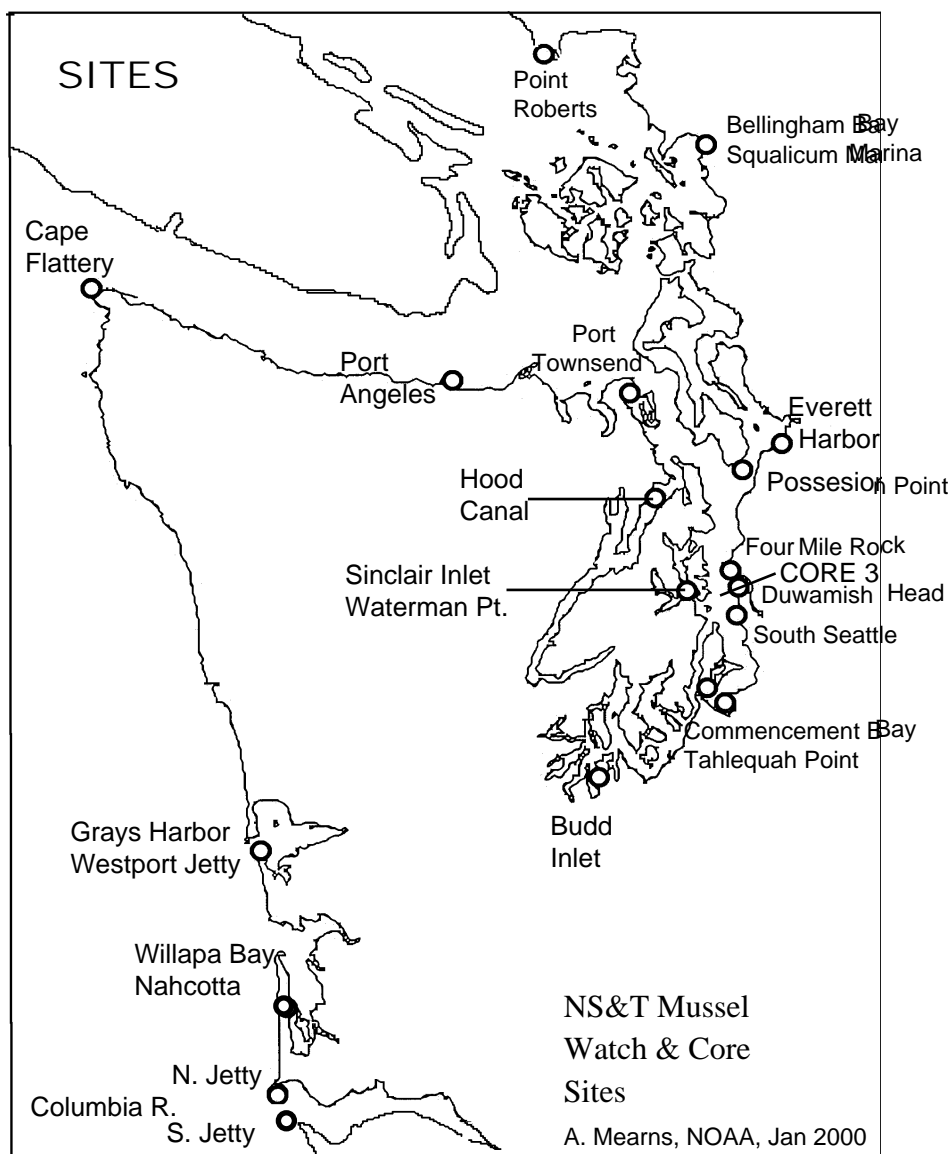


Figure 1 Names and locations of National Status and Trends Mussel-watch Sites in Washington and at the Columbia River, Oregon. Also noted is the location of Core #3 in Puget Sound.

Dated Sediment Cores

In 1990 Lefkovitz and others (1997) carefully took meter-long cores from six sites in the deepest parts of the Main Basin of Puget Sound. The cores were sectioned and the sections dated using geochemical methods and analyzed for all the chemicals described above. Depending on core depth, dates ranged from the mid-1800s to about 1990, thus slightly overlapping the Mussel Watch sampling period by several years. Lefkovitz and others (1997) published the data as vertical profile plots (distance down core and time on the vertical axis.) Time and resources did not permit the author to conduct a full review of all six cores. Core 3, near Elliot Bay and the Four Mile Rock mussel watch site, probably represents the most historically contaminated of the six sites. For this paper I extracted the data only for Core 3 and replotted them placing time on the horizontal axis and chemical concentrations on the vertical. This resulted in graphics comparable to other plots of time series, such as the Mussel Watch data and numerous comparable time series presentations such as for fishery statistics (West and others 1997), benthic infauna trends (Nichols, 2001) and other historical pollution and resource trends (eg., as in Dexter and others 1984). Hopefully this presentation will serve as a model for others to complete.

Units

All concentration data for sediments and mussels are in units of dry weight: mg/kg dw (ppm) or ug/kg dw (ppb). In the text and graphics that follow these concentrations are referred to as ppm dw or ppb dw. Seafood guidelines are generally presented on a wet or fresh tissue weight basis. Mussels generally contain about 80% water so to obtain a rough estimate of wet weight concentrations for contaminants in mussels the reader should divide given values by 5.

Results

Sites and Site Groupings

Table 1 summarizes information about the 17 Washington NS&T Mussel Watch sites and one Columbia River South Jetty (Oregon) site. These are also identified in Figure 1. Site names were assigned in 1986 and do not necessarily reflect local usage. The sites at Port Townsend, Port Angeles and Hood Canal were assigned to the general location category "Puget Sound" by the national program but the first two are on the Straits of Juan de Fuca. The site "Tahlequah Point" is not in Commencement Bay but north and opposite of the Bay near the ferry terminal at the southern end of Vashon Island. Specific collecting stations, and instructions for locating them are given in Lauenstein and others (1997). Effort was made to locate sites away from known point sources and also to correspond to sites sampled during the EPA National Mussel Watch of the late 1970s (Lauenstein and Daskalakis 1998). Collections are from rocky or gravel beaches but some collecting locations include rip rap jetties and pilings either away from marinas (Columbia River, Westport) or near them (Port Townsend Marina, Squalicum Marina.) Counties are also assigned in Table 1 to help Marine Resource Committees (MRCs) identify sites in their counties. For the purpose of this paper, sites are also group by general area as shown in Table 1. For example, "Coast" refers to all sites between the Columbia River South Jetty and Cape Flattery.

Description of Graphics

The maps in Figures 2 and 3 show the 1997 through 1998 concentrations of 9 trace elements (Figure 2) and 8 groups of organic chemicals and mussel lipid content (Figure 3). Each map also shows the complimentary statistics for the entire U.S. population of mussels (n=123 for metals) or mussels and oysters (n = 252 for organic chemicals) collected in 1997 and/or 1998. Both mussels and oysters are sampled nationwide. The NS&T Mussel Watch Program has determined that oysters accumulate much higher concentrations of copper and zinc than do mussels while no such differences exist for other chemicals (O'Connor 1998). To be conservative, Washington Mussel Watch data for metals are compared only to the US mussel collections that include 123 sites nationwide.

The maps in Figures 2 and 3 are complimented by scatter diagrams showing trends for all the data from 1986 through 1998 in Figures 4 and 5. All replicate values are shown for all stations where data was produced. Because of the small size of these plots it is only possible to resolve sites with extreme concentrations. There is an appearance in these plots of less data after 1992: this reflects the reduction from three to one composite per station and a reduction in sampling frequency from annual to biennial.

Long-term (approximately 1890 to 1990) time series plots for 15 metals, total organic carbon (TOC), nitrogen (N) and phosphorus (P) in sediments from Lefkovitz and others (1997) Core 3 near Seattle are shown in Figure 6 as deviations from concentrations that prevailed in the 1890's: presumably these were at or near the actual background concentrations for Puget Sound. This plot allows a look at those contaminants that increased by more than about 50% above background (1.5 times) but does not easily resolve those that increased below this factor. The actual (non-normalized) values are given in Lefkovitz and others (1997). Figure 7 is a composite of comparable century-long trends for 7 selected organic chemicals or chemical groups. Values for nitrogen and phosphorus are also included.

These plots (Figures 1 through 7) form the basis of the individual chemical narratives that follow Figures 2 through 6 on the next pages.

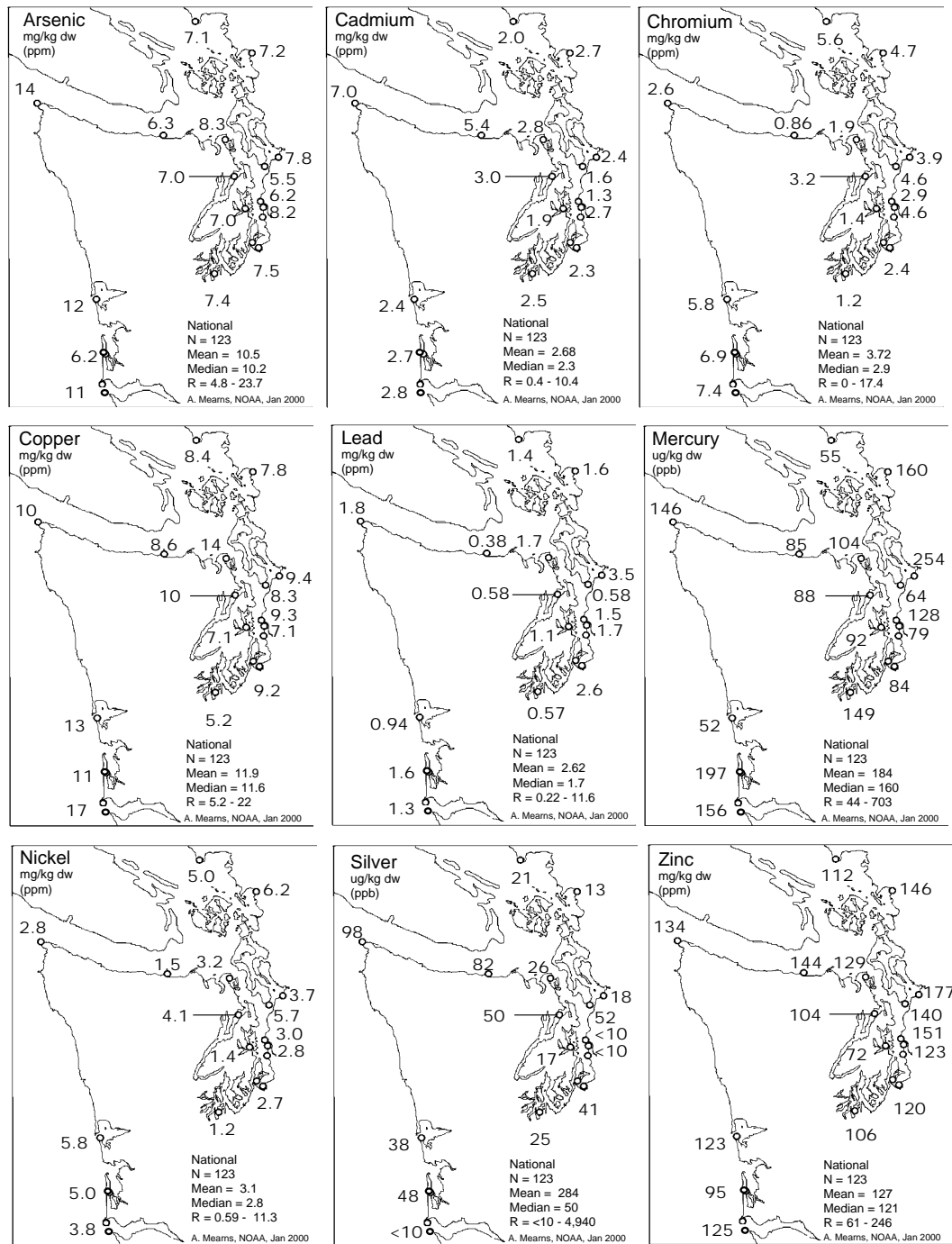


Figure 2. Concentrations of nine trace elements in mussels collected from Washington coastal sites in 1997 or 1998. Also included are statistical summary of National data.

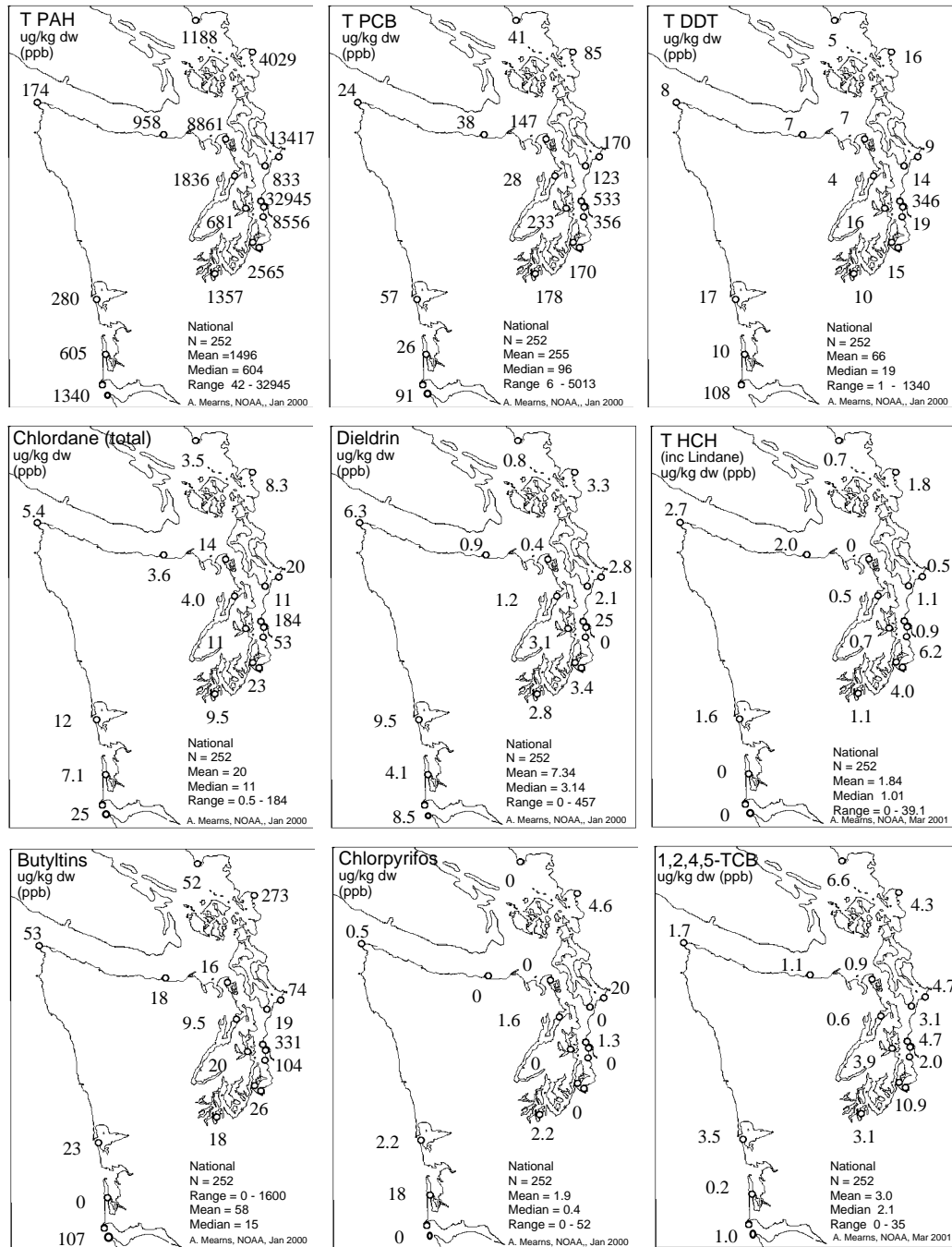


Figure 3. Concentrations of 9 organic contaminants in mussels collected from Washington coastal sites in 1997 or 1998. Also included are statistical summaries of national data.

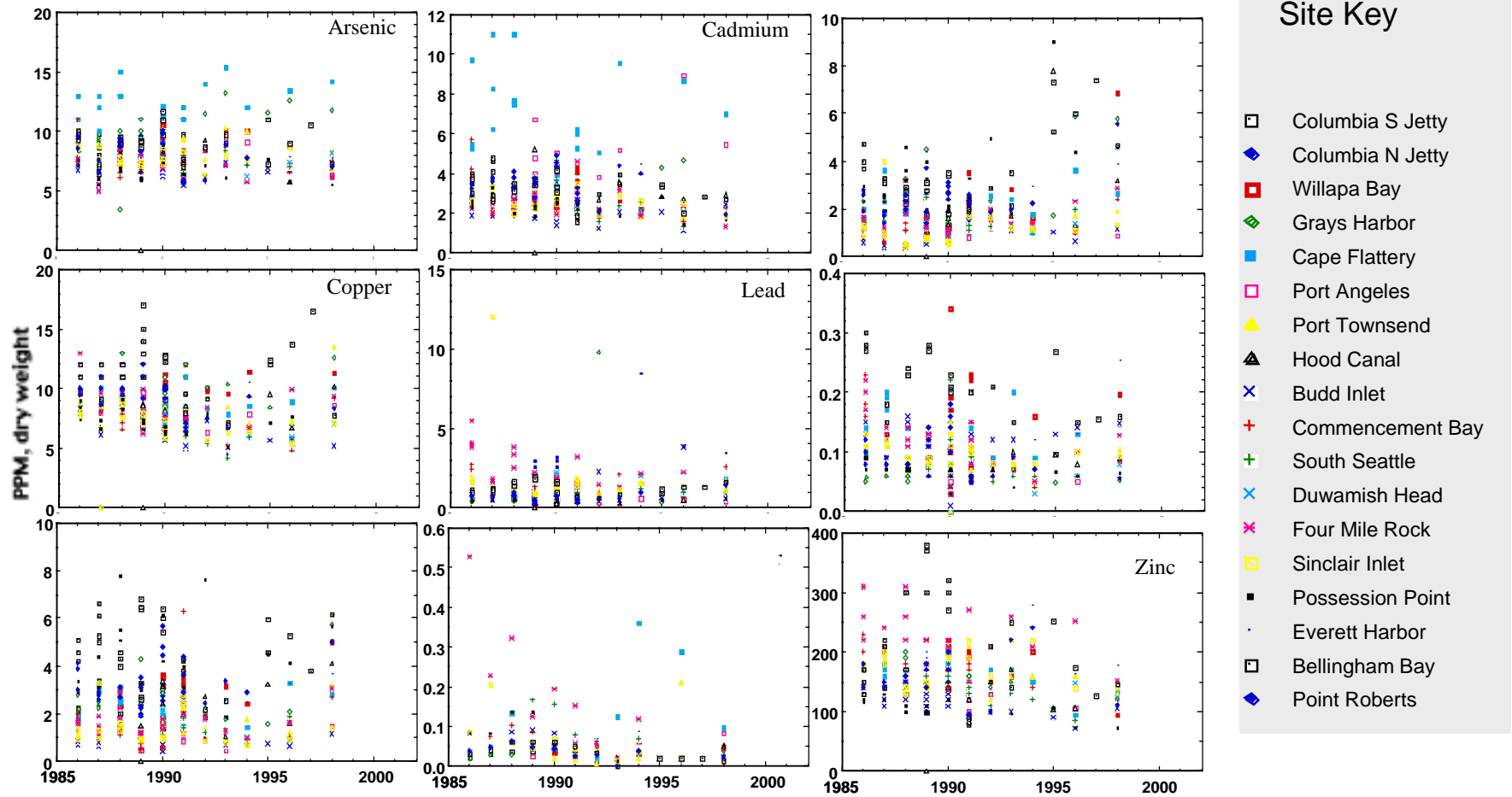


Figure 4. Time series trend of concentrations of 9 trace minerals in mussels from 18 National Mussel Watch sites in Washington and Oregon.

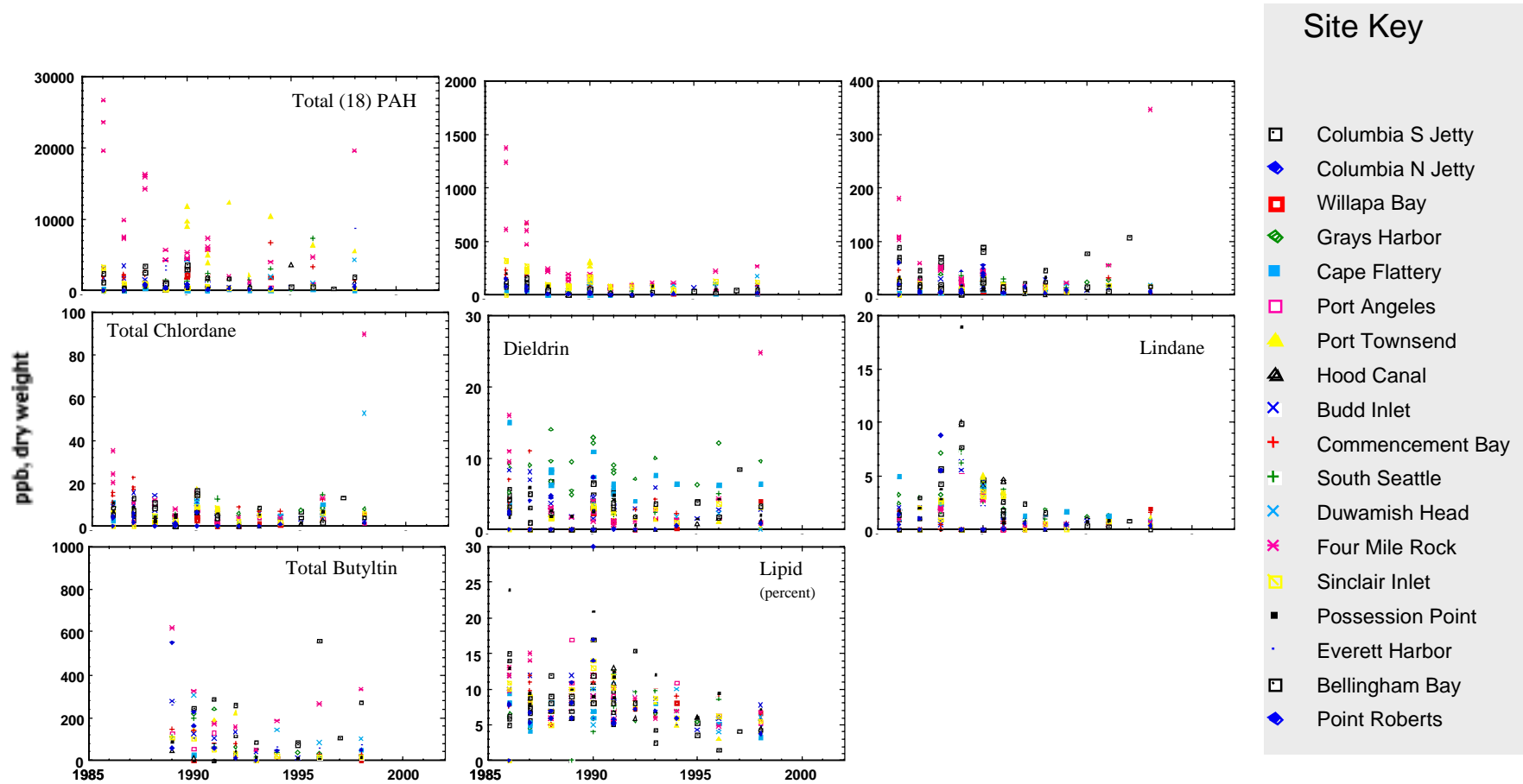


Figure 5. Time series trends of concentrations of 7 organic chemical groups in mussels from 18 National Mussel Watch sites in Washington and Oregon.

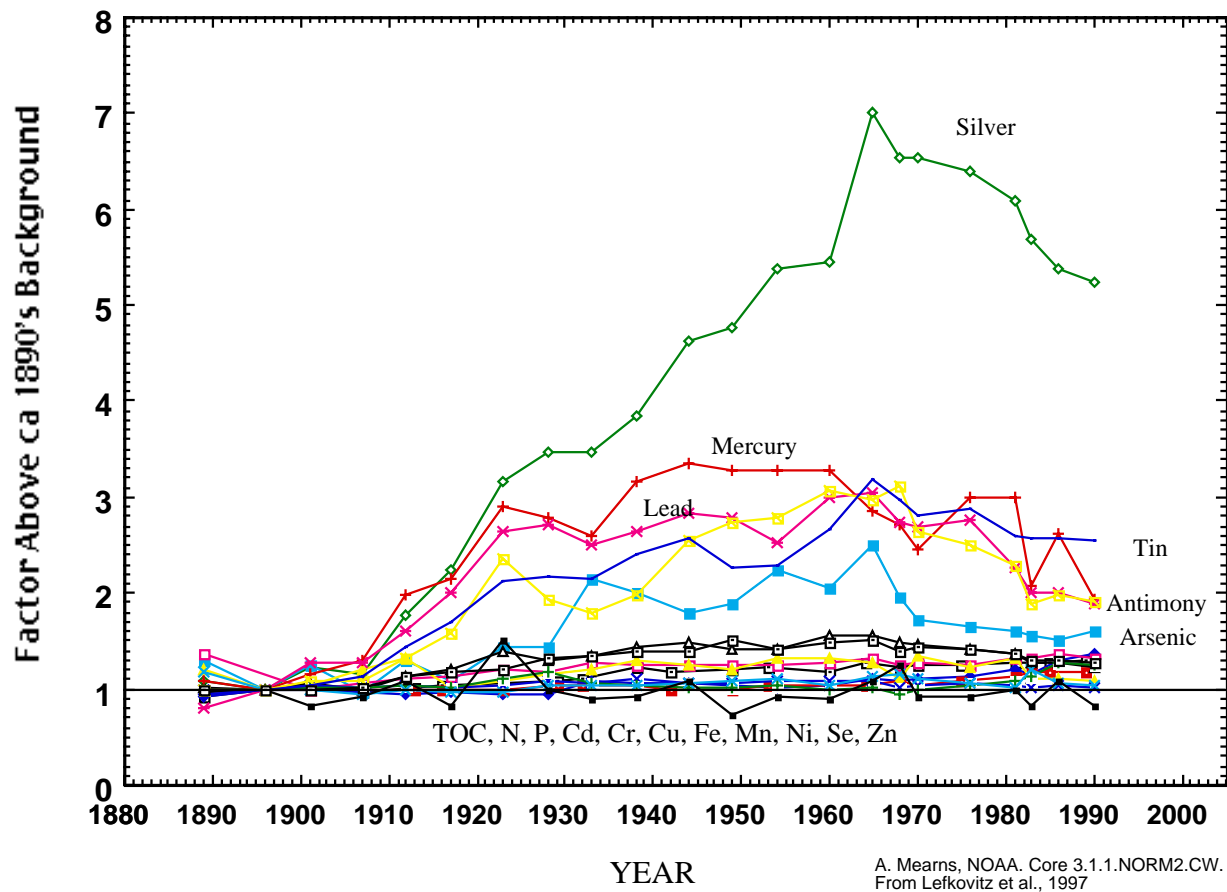


Figure 6. Trends in the concentrations of 17 metals and conventional materials, 1880 to 1990, in dated Core #3 from central Puget Sound based on data in Lefkovitz and others (1997). Values are ratios of given-year concentrations to concentrations prevailing in approximately 1895.

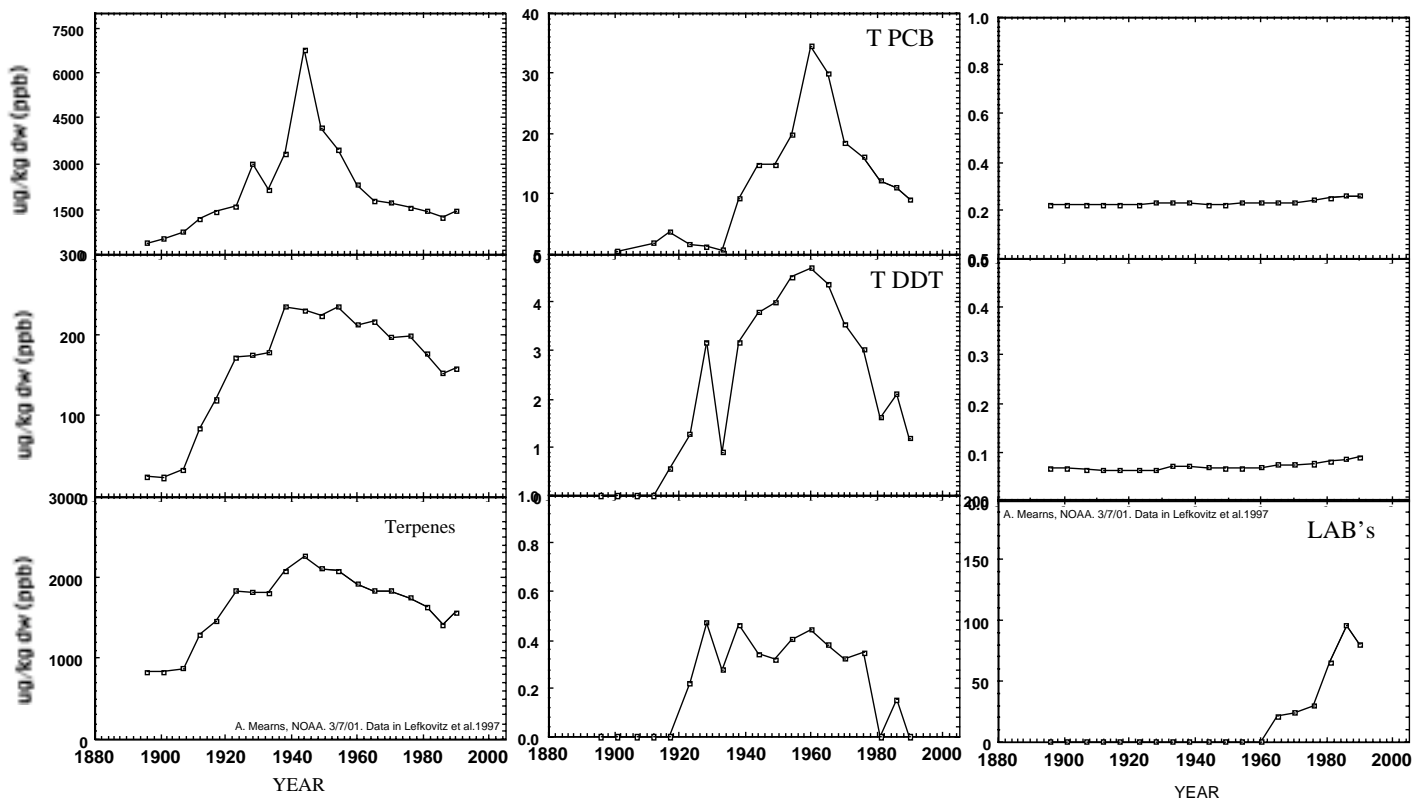


Figure 7. Time series trends of concentrations of eight contaminant or material groups in dated Core 3 from central Puget Sound. Based on data in Lefkovitz and others 1997.

Trace Metal Patterns and Trends

Arsenic (As) concentrations in Washington mussels in 1998 ranged about threefold from 5.5 ppm dw at Possession Point (Whidbey Island) to 14.0 ppm at Cape Flattery (Figure 2). Concentrations were higher on the coast (6.2 to 11 ppm) than in Puget Sound (5.5 to 7.8 ppm) and the Northwest Straits (6.3 to 8.3 ppm). The three highest concentrations were from coastal samples of the California mussel, *Mytilus californianus*. With the exception of Cape Flattery, Washington mussels generally contained lower concentrations of arsenic than the national mean of 10.5 ppm dw (range 4.8 to 23.7 ppm dw). There were no obvious long-term trends in arsenic from 1986 to 1998 in the collective data from Washington (Figure 4); however, there does appear to be a cyclic pattern with five year peaks at Cape Flattery (highest concentrations, Figure 4). In Core 3 from Puget Sound arsenic concentrations began to increase above background between 1910 and 1930 reaching peak concentrations of 2 to 2.5 times background in the 1960s and then declined to concentrations about 15 times background by about 1990 (Figure 6).

Cadmium (Cd) concentrations in mussels collected in 1998 also ranged about threefold from a low of 1.3 ppm dw at Four Mile Rock in Elliot Bay to 7.0 ppm dw at Cape Flattery (Figure 2.) The second highest concentration, 5.4 ppm dw, came from mussels collected at Port Angeles. By contrast, the second lowest concentration, 1.6 ppm, came from mussels inside Puget Sound at Possession Point. Cadmium concentrations in Washington mussels were similar to the national mean of 2.66 ppm dw and well within the national range of 0.4 to 10.4 ppm dw. As shown in Figure 4, and with notable exception of Cape Flattery, there appears to be a general trend of decreasing concentrations of about 1 ppm for all the Washington sites taken as a group. This hypothesis needs to be tested statistically. During the past century cadmium concentrations in sediments of Puget Sound Core 3 did not appear to increase beyond a factor of 1.1-1.3 times the 1890s background.

Chromium (Cr) concentrations in 1998 mussels ranged about sevenfold from a low of 0.86 ppm dw at the station in Port Angeles to 7.4 ppm in mussels from the Columbia River South Jetty (Figure 2). Coastal concentrations of chromium were higher than those in mussels sampled from the interior regions of the Sound and Northwest Straits. The Washington chromium concentrations were comparable to the national average of 3.72 ppm and range of 0 to 17.4 ppm. Chromium concentrations at four or five sites appeared to suddenly increase between 1994 and 1995 (Figure 4); this may be an analytical artifact as described in O'Connor (1998). Aside from this, it also appears that there was a modest (1 to 2 ppm) decrease in chromium concentrations during the 1990s, a trend requiring statistical confirmation. During the 100 years between the 1890s and 1990s chromium was apparently not a major contaminant in the Main Basin at the Core 3 site: concentrations never increased above a factor of 1.3 to 1.5 background (Figure 6).

Copper (Cu) concentrations in 1998 mussels ranged threefold from 5.2 ppm dw in Budd Inlet to 17 ppm dw at the Columbia River South Jetty (Figure 2). The highest concentrations were in mussels from the coast (range 10 to 17 ppm dw.) Copper concentrations from mussels in the Northwest Straits and Puget Sound were all below the national average of 11.9 ppm dw and at the low end of the national range of 5.2 to 22 ppm dw, whereas those from the coast were equal to or above the national average. Mussel copper concentrations displayed no systematic trend between 1986 and 1998 (Figure 4); however, there did appear to be a general cycle with higher concentrations in 1989 and 1996-98 and lower concentrations in 1986 and the early 1990s. Copper was apparently not a substantial historic sediment contaminant in central Puget Sound; sediment concentrations never exceeded 1.3 to 1.5 times the 1890's background, reaching this peak in the 1950s and 1960s and declining since then (Figure 6).

Lead (Pb) concentrations ranged about tenfold, from 0.38 ppm dw at Port Angeles to 3.5 ppm dw at Everett Harbor (Figure 2). With the notable exception of Everett Harbor, the ranges of lead were similar in mussels from the coast, the Northwest Straits and Puget Sound. Washington mussel lead concentrations were generally below the national average of 1.7 ppm dw and at the lower end of the national range of 0.22 to 11.6 ppm dw (Figure 2). There does appear to be a secular trend of decreasing lead concentrations in Washington mussels between 1886 and 1998 (Figure 4). Closer inspection of the data (not shown) indicates mussels from stations near Seattle were clearly contained higher concentrations in 1986 than in 1998. The high values for lead in Figure 4 may be an artifact: in past years when lead was a very common atmospheric pollutant it was very difficult to avoid lead contamination of environmental samples.

Historically, lead was clearly a substantial contaminant in sediments from the Main Basin of Puget Sound: Lefkovitz and others (1998) Core 3 experienced rising concentrations from about 1910 to 1920 with levels nearly 3 times background from about 190 to 1980 followed by a decline to about 1.7 times background in 1990 (Figure 6).

Mercury (Hg) concentrations in 1998 Washington mussels ranged fivefold from 52 ppb dw at the Westport Jetty to 254 ppb dw at the Everett Harbor site. With the exception of Everett harbor, the range of mercury concentrations was similar in mussels from coastal sites (52 to 197 ppb dw), the Northwest Straits (85 to 160 ppb dw) and Puget Sound (64 to 149 ppb dw). Most of the concentrations in Washington were below the national mean of 184 ppb dw and toward the low end of the national range of 44 to 703 ppb dw. There also appeared to be a trend of decreasing mercury concentrations in Washington mussels from 1988 to 1996 and then a leveling-off in 1998 (Figure 4). The highest concentrations in the 1980s were at the Bellingham site but these appear to have declined to within the range of Washington concentrations by 1998. The dated sediment Core 3 clearly indicates that mercury was a substantial sediment contaminant in the Main Basin: concentrations began to increase in 1905-1910 rising to values over 3 times background from about 1940 to 1960 and then decreasing to less than 2 times background by 1990 (Figure 6).

Nickel (Ni) concentrations in 1998 Washington mussels ranged less than threefold from a low of 1.2 ppm in mussels from Budd Inlet to a high of 6.2 ppm at the Squalicum Marina Jetty in Bellingham. Ranges of concentrations were generally comparable in mussels from coastal sites (2.8 to 5.8 ppm dw), the Northwest Straits (1.5 to 6.2 ppm dw) and Puget Sound (1.2 to 5.7 ppm dw). There was apparently no secular trend in nickel concentrations between 1986 and 1998; however, during the period 1992-1994 concentrations were substantially lower than either before or after (Figure 4). Puget Sound dated Core 3 apparently did not experience substantial nickel contamination (Figure 6).

Silver (Ag) concentrations in 1998 Washington mussels ranged at least tenfold from lows of < 10 ppb dw at Four Mile Rock and Duwamish Head in central Puget Sound, and the Columbia River South Jetty, to a high of 98 ppb dw in mussels from Cape Flattery. All silver concentrations in Washington mussels were far below the national average concentration of 284 ppb dw (range < 10 to 4,940 ppb dw.) . Although there appears to be a tendency for higher silver concentrations in mussels from the coast compared to the Straits or the Sound, there were no clear regional differences with one exception: silver was not measured above the detection limit of 10 ppb dw at the two sites near Seattle. These observations are in stark contrast to the long-term history of silver in Main Basin sediments: As shown in Figure 6 silver was the only trace metal that reached sediment concentrations exceeding 4 time historical background concentrations. Main Basin sediment silver concentration began to increase in the early 1900s, reaching twice background by 1910, three-time background by 1920, five-times by 1945 and seven-times by 1960. At this time silver inputs apparently began decreasing such that by 1990 surface sediment concentrations were at or below six times background.

Zinc (Zn) concentrations in 1998 Washington mussels ranged only twofold, from a low of 72 ppm dw at Waterman Point, Sinclair Inlet to 177 ppm in the Everett Harbor mussels. With the exception of the Sinclair Inlet site, zinc concentrations appeared to be comparable among the three geographic areas. Concentrations of zinc in Washington mussels were within the national range of 62 to 246 ppm dw. As shown in the last panel in Figure 4, zinc concentrations in Washington mussels appear to have decreased substantially between 1986 and 1996. The collective and individual site trends should be analyzed statistically. Despite this, zinc did not appear to be a substantial historical contaminant in sediments from the main Basin: concentrations never exceed 1.3 to 1.5 times background (Figure 6).

Organic Chemical Patterns and Trends

Total PAHs in 1998 Washington mussels ranged over fiftyfold, from a low of 174 ppb dw at Cape Flattery to a high of nearly 33,000 ppb in the mussels from Four Mile Rock in Elliot Bay. Unlike metals, there appeared to be strong geographical gradients for PAHs (such as a 30-fold decrease from Seattle to Budd Inlet, or from the Columbia River north to Cape Flattery, Figure 3) as well as possible local "hot spots" (Everett Harbor, 13417 ppm dw; Port Townsend, 8861 ppm dw; Figure 3). Significantly, the highest TPAH concentration in the 1997-98 mussel watch program in the US was at Four Mile Rock. T PAH's in mussels

from several urban sites also experienced dramatic decreases, especially during the period 1986 to about 1994: after this, T PAH's increased at several sites, notably at Four Mile Rock (Figure 5). PAHs have been notable contaminants of Main Basin sediments throughout the past century. Concentrations increased from about 300 ppb dw in 1990 to over 7000 ppb dw by about 1945, and then decreased to about 3000 ppb dw by 1960 and 1500 ppb dw by 1990 (Figure 7). If a value of about 300 ppb dw can be taken as a natural sediment background, then the bottom sediments of Puget Sound experienced concentrations 25 to 30 times background during the World War II years, decreasing to 5 time background by 1990.

Total PCB concentrations in 1998 Washington mussels ranged twentyfold from a low of 24 ppb dw at Cape Flattery to a high of 533 ppb dw at Four Mile Rock in Elliot Bay (Figure 3). Concentrations throughout Puget Sound (123 to 5533 ppb dw) were considerably higher than in the Northwest Straits (28 to 147 ppb dw) or the coast (26 to 91 ppb dw, Figure 3). Despite this, T PCB's in Washington mussels were generally at or below the national mean (252 sites, both mussels and oysters) of 255 ppb dw (range 6 to over 5000 ppb dw). Indeed, the median for Puget Sound (178 ppb dw) was below the National average. T PCBs were substantially higher in mussels 1986 than in recent years (Figure 5); for example, average concentrations at Four Mile Rock were over 1000 ppb dw in 1986 (Figure 5). Concentrations in most areas decreased substantially during the 1980's. However, it also appears that they have stopped decreasing and possibly started increasing again during the mid 1990's (Figure 5). Historically, T PCBs have been substantial contaminants in sediments of the Main Basin since the 1930s; as shown in Figure 7, concentrations increased from nearly zero about 1930 to 35 ppb dw by 1960. Then inputs apparently decreased dramatically, with sediment concentrations dropping below 15 ppb dw in 1980 and less than 10 ppb dw in 1990 (Figure 7).

Total DDT concentrations in 1998 Washington mussels ranged nearly hundredfold from a low of 4 ppb dw in Hood canal to a high of 346 ppb dw in mussels from Four Mile Rock (Figure 3). The second highest concentration, 108 ppb dw, occurred in mussels from the Columbia River South jetty. With these two exceptions, T DDT concentrations in Washington mussels ranged from 4 to 20 ppb dw with levels generally comparable among the three geographic areas. These concentrations are close to the national median of 19 ppb dw but well below the national mean of 66 ppb dw (range 1 to 1340 ppb dw; Figure 3). It appears that T DDT concentrations were decreasing in Washington mussels when this monitoring program began in 1986 (Figure 5). However, there was a sharp increase at Four Mile Rock in 1997-98 sampling. Like PCB's, T DDT was a substantial historical contaminant in Main Basin sediments: concentrations began increasing in the 1930's reaching a peak of nearly 5 ppb dw in 1960 and then decreasing to about 1 ppb dw in 1990 (Figure 7).

Total Chlordane concentrations (sum of 7 analytes including heptachlors) in 1998 Washington mussels ranged about fiftyfold from a low of 3.5 ppb dw at Point Roberts to 184 ppb dw in mussels from the Four Mile Rock site (Figure 3). This site also produced the highest total chlordane concentration in the US in the 1997-98 mussel watch surveys. Other Washington sites were generally within a factor of 2 of the 1997-98 national mean of 20 ppb dw. From Figure 5 it is apparent that chlordanes (4 compounds) were decreasing in Washington mussels when the program started in 1986, and continued to do so through about 1995. However, there appeared to be an increase in 1996 and definitely so for several sites that were sampled in 1997-98. That increase occurred at Four Mile Rock and is responsible for the high value reported above. Chlordanes were not analyzed in the sediment cores.

Dieldrin concentrations in 1998 Washington mussels ranged from below detection (0) at South Seattle (Fauntleroy), and 0.4 ppb dw at Port Townsend to 25 ppb dw in mussels from Four Mile Rock (Figure 3). Aside from this site, concentrations in Puget Sound and in the Northwest Straits (range 0.4 to 3.3 ppb dw) were considerably lower than in mussels from all four coastal sites (range 4.1 to 9.5 ppb dw, Figure 3) and were within a factor of two of the national mean of 7.34 ppb dw (range 0 to 457 ppb dw). Inspection of Figure 5 suggests dieldrin concentrations were declining when the monitoring began in 1986 and continued to do so until a sharp rise at several sites in 1997-98. Dieldrin was a measurable contaminant in main Basin sediments from 1925 to 1960 (Figure 7).

T HCH compounds, including **lindane**, in 1998 Washington mussels ranged zero at Port Townsend, the Columbia River and Willapa Bay to 6.2 ppb dw at South Seattle (Fauntleroy, Figure 3); a value of 4 ppb

dw was also recorded at Tahlequah Point near Commencement Bay. It is also of interest to note that the third highest concentration (2.7 ppb dw) occurred at Cape Flattery. With the exception of South Seattle and Tahlequah Washington HCH concentrations were within a factor of two of the 1997-98 national mean of 1.84 ppb (range 0 to 39.1 ppb dw, Figure 3). Lindane itself has been monitored since the inception of the NS&T program. Curiously, concentrations in Washington mussels increased from 1986 to 1989 and then decreased (Figure 5). Lindane or HCH was not measured in the sediment cores.

Butyltin compounds, including tributyltin, in 1998 Washington mussels dramatically ranged from zero in mussels from Willapa Bay to 331 ppb dw (Figure 3). The highest concentrations occurred at Four Mile Rock (331 ppb dw), Squalicum Marina Jetty in Bellingham (273 ppb dw) and at the Columbia River South Jetty (107 ppb dw, Figure 3). It is also curious that the mussels at Cape Flattery had a higher concentration (53 ppb dw) than those from Port Angeles, Port Townsend, Hood Canal, Sinclair Inlet, Tahlequah Point and Budd Inlet (range 9.5 to 26 ppb dw). Generally, Washington mussel concentrations of butyltins were at or below the 1997-98 national mean of 55 ppb dw and also at the low end of the national range (0 to 1600 ppb dw). As shown in Figure 5, it appeared that total butyltin concentrations were decreasing when the mussel watch program began in 1986, and continued to do so until 1996-98 when several sites experienced increases. Butyltins were not included in the sediment core analytes reported by Lefkovitz and others (1997).

Chlorpyrifos was undetected at half the Washington sites sampled in 1998 (Figure 3). However, there were three substantial concentrations: 20 ppb dw at the Everett Harbor site, 18 ppb dw at Nahcotta in Willapa Bay and 4.6 ppb dw at the Squalicum Marina Jetty in Bellingham (Figure 3). These concentrations were above the 1997-98 national mean of 1.9 ppb dw (median 0.4 ppb dw, range 0 to 52 ppb dw). These concentrations are comparable to the highest concentrations found elsewhere in 1994-97 (Wade and others 1998). Chlorpyrifos was not measured in the dated sediment cores.

1,2,4,5-tetrachlorobenzene and other chlorobenzenes have recently been measured in mussels by the NS&T program. As shown in Figure 3, concentrations of this compound in 1998 Washington mussels ranged from 0.2 ppb dw at Nahcotta, Willapa Bay, to 10.9 ppb dw at Tahlequah Point near Commencement Bay. The second highest concentration was 6.6 ppb dw at Point Roberts. Concentrations in mussels from the Sound were higher than in the other regions whereas those in the Northwest Straits and along the coast were comparable. There is not yet enough time series data to develop a picture of long-term trends and these compounds were not measured in dated sediment cores.

Mussel **tissue lipids** were also examined for trends because they can control total dry weight concentrations of the organic contaminants. In 1998 percent lipids were slightly (1.3 times) higher in mussels from Puget Sound than in those from the Northwest Straits or the coast (data not shown). Also of some interest is the possibility that Washington mussel lipid concentrations have decreased noticeably in the 1990s following a previous period of stable but higher concentrations (Figure 5).

The chemical markers **linear alkyl benzenes (LABs)**, an index of modern detergents), **hopane** (a non-biodegradable index of petroleum) and **terpenes** (natural wood product) were measured in dated cores by Lefkovitz and others (1997) but not in mussels by the NS&T program. As shown in Figure 7, there is a long history of hopane and terpene contamination in Puget Sound Main Basin Core 3, with peak inputs apparently occurring in 1940 and decreasing thereafter. These may be indices of trends in oil spills (hopane) and paper industry byproduct discharges (terpenes). Of all the chemicals measured in cores or mussels, **LABs** appear to be the only ones undergoing increasing concentrations (beginning in the 1960's, Figure 7).

Also shown in Figure 7 are century-long trends in total **nitrogen (N)** and **phosphorus (P)** in Main basin Core 3. In short, the curves are almost flat: a slight and significant increase is apparent but there is no dramatic trend comparable to those measured for the other contaminants and materials.

Discussion

The major goal of this work is to stimulate the continued study of long-term, region-wide contaminant trends in Puget Sound and the Northwest Straits. Although the Mussel Watch and sediment core data have been previously published, they have not been brought together in a form useful for telling stories about long-term and recent trends or for comparing geographic areas or comparing conditions in Puget Sound with the rest of the U.S. coast. The NOAA Mussel Watch Program provides those perspectives while the dated core project provides a sense of the long history of contamination and recovery.

This brief review uncovered interesting patterns and trends that have not, to my knowledge, been previously explored. Below, I point out just a few of the patterns and, at some risk, speculate on what they might mean.

Geographic Patterns and Anomalies

From a regional geographic point of view it is interesting to discover that, for the most part, contemporary (1998) concentrations of chemicals in Washington mussels are comparable to those nationally. For example, I was surprised to learn that local and regional concentrations of PCBs, while presumably of concern, were at or below the “national average.”

Within Washington there are several important contaminant patterns that when viewed regionally suggest priorities for concern. To explore these further I computed mean analyte concentrations for the three geographical groups described in Table 1 and compared Puget Sound and Northwest Strait means to those from the coast. As shown in Table 3, only three contaminant groups at the Northwest Straits sites exceeded comparable concentrations on the coast (ie, the ratio of means was > 1): PAHs were over 4 times higher, pentachlorobenzene about 3 times higher and selenium slightly higher (1.2 times). Mean concentrations of 8 materials were higher in Puget Sound than on the coast (PAHs, 5.8; PCBs, 4.3; a chlorobenzene 2.7; total chlordanes, 2.1, etc). Curiously mean lipid concentrations in the Puget Sound mussels were also tended to be higher (1.2 times) than on the coast.

However, in remarkable contrast is the observation that at least a dozen materials that we regard as contaminants appeared to be lower in either the Northwest Straits or Puget Sound than in coastal mussels (mean ratios < 1 ; Table 3). For example, nearly two-thirds of the trace metals appeared in lower concentration in coastal mussels than in those from the Straits or Puget Sound. Conversely, one could conclude that mussels in the Sound are depleted in trace metals compared to those on the coast. Such a situation may be normal or an anomaly. Some trace elements, such as mercury, arsenic and cadmium may be elevated naturally in mussels living near ocean upwelling areas (reviewed in Mearns and others 1991) or near seal and sea lion colonies (eg., mercury, Flegal and others 1981). Alternatively, high tissue concentrations of organic contaminants (such as PAHs, PCBs, DDTs) could cause mussels and other animals to experience abnormalities in trace element metabolism. For example, depressed metals were found in the livers of diseased Southern California flatfish highly contaminated with DDT and PCBs: likewise, we also found a significant inverse relation between cadmium and T DDT in mussels and oysters from throughout the U.S. (both situations reviewed in Mearns and others 1991). In any case, such situations bring into focus, at least for trace metals, the question, “how clean is clean enough?” and beg that contaminants not be examined in isolation of one another or in isolation of oceanographic patterns and trends.

Several contaminants presented themselves in isolated locations. In 1998 the current-use pesticide chlorpyrifos appeared at relative high concentrations at Nahcotta (Willapa bay) and also at Everett Harbor. Its appearance in Willapa Bay is consistent with its use in cranberry culture operations there (Art Johnson, WDOE, Personal communication, February, 2001). Perhaps there is also local use of this pesticide in the Snohomish River drainage as well.

Table. 3. Ratios of region-to-coast median contaminant concentrations in mussels for sites in the Northwest Straits and sites in Puget Sound. N = 4 for the coastal region. For ratios > 1, regional median is higher than coastal. See Table 1 for regional station groupings. Based on NOAA NS&T Mussel Watch data for 1997-98.

Northwest Straits (N=5)			Puget Sound (N=7)		
Rank	Analyte	Ratio	Rank	Analyte	Ratio
1	Total PAHs	4.1	1	Total PAHs	5.8
2	Pentachlorobenzene	3.0	2	Total PCBs	4.3
3	Selenium (Se)	1.2	3	T1245CB	3.0
4	Zinc (Zn)	1.0	4	Pentachlorobenzene	2.7
5	Cadmium (Cd)	1.0	5	Total Chlordane (7)	2.1
6	Total PCBs	1.0	6	Total HCH	1.3
7	Lead (Pb)	1.0	7	Lipid (%)	1.3
			8	T DDT	1.1
8	Nickel (Ni)	0.9	9	Lead (Pb)	1.0
9	Lipid (%)	0.9	10	Zinc (Zn)	1.0
10	Manganese (Mn)	0.8	11	Manganese (Mn)	1.0
11	Total HCH	0.8	12	PP DDE	1.0
12	T1245CB	0.8			
13	T1234CB	0.7	13	Selenium (Se)	0.8
14	Copper (Cu)	0.7	14	Cadmium (Cd)	0.8
15	Arsenic (As)	0.6	15	Copper (Cu)	0.7
16	Silver (Ag)	0.6	16	Total Butyltins	0.7
17	Mercury (Hg)	0.6	17	Arsenic (As)	0.7
			18	Nickel (Ni)	0.6
18	Aluminum (Al)	0.5	19	Mercury (Hg)	0.6
19	T DDT	0.5			
20	Chromium (Cr)	0.5	20	Tributyltin	0.5
21	Total Butyltins	0.5	21	Chromium (Cr)	0.5
22	Total Chlordane (7)	0.4	22	Silver (Ag)	0.4
23	PP DDE	0.4	23	Aluminum (Al)	0.4
24	Iron (Fe)	0.4	24	Dieldrin	0.4
25	Tributyltin	0.3	25	Iron (Fe)	0.3
26	Dieldrin	0.1	26	T1234CB	0.3
27	Chlorpyrifos	0.0	27	Chlorpyrifos	0.0
28	Tin (Sn)	-	28	Tin (Sn)	-

Long-Term Trends

Two scales of temporal trends—100 years and about a dozen years—can be explored in the data partially examined for this report. Contaminants in the cores, dated and analyzed by Lefkowitz and others (1997), provide a pre-industrial background showing what the chemical profiles looked like in the 1880s and 1890s. Trace metals, nitrogen, phosphorus, PAHs, and indicators of petroleum (hopane) and wood (terpenes) were present and at easily measured concentrations which could be taken as natural background levels for purposes of defining “how clean is clean.” Pesticides, PCBs, and other chlorinated chemicals were not present. These materials appeared beginning in the 1920s and increased to peak concentrations in the 1940s to 1960s after which they began to decrease. The rates of increase and decrease should be proportional to their local mass inputs from all sources. During these “peak” years there were also modest increases in concentrations of most trace elements, but five—silver, mercury, lead, tin, antimony and arsenic—experienced dramatic increases (ie greater than a factor of 2 above “background,” Figure 7. The most dramatic increase was for silver, increasing in sediments off Seattle (Core 3) to concentrations over 7 times background by 1965. Using data from the California Mussel Watch Martin and others (1988) speculated that silver was a useful marker of sewage contamination. Presumably either pre-treatment, source control or primary sewage treatment (or all three) in Puget Sound were sufficient during the 1960s to dramatically reduce silver inputs to the Sound. However, there remains an anomaly: silver was not only

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not a substantial contaminant in mussels in Puget Sound during the late 1980s and 1990s but it was actually depleted in mussels from the two Seattle stations located in close proximity to Core 3 where, in 1990, deep basin sediment concentrations were still 5.5 times background. Thus either the silver was not biologically available or some other factor was inhibiting “normal” silver accumulation (such as at Cape Flattery) in these mussels, or both.

Likewise, despite the fact that mercury, lead, tin and arsenic were still elevated several times above background in Puget Sound Main Basin sediments in 1990, no such elevations were observed in the mussels. The possibility exists that trace metals, whether natural in origin or from anthropogenic sources, are well sequestered in the sediments and are (or were) not available for bioaccumulation.

The dated core data suggest that during and following the World War II years, PCB concentrations (and, presumably, inputs) were 4 to 5 times higher than they have been during the 1990s. Such trends have implications for understanding the role contaminants may or may not be playing in determining the health of marine life in the Sound. Our fish and local seafood, which apparently were much more abundant 30 to 40 years ago than now, were presumably 4 to 5 times more contaminated with PCBs than they are today. Indeed, while concentrations of PCBs and other contaminants were declining during the past third-century, the abundances of key fish species were also declining. If we are concerned about the effect of current concentrations of these contaminants on fish reproduction now, we should consider the possibility that reproduction was at least 10-times more impacted in the mid 20th century.

In contrast to these patterns is the complete lack of comparable trends for sediment nitrogen and phosphorus. Despite presumably heavy inputs of organic material from pulp mills during the 1950s and 1960s, and from large wastewater treatment systems in the 1970s (Dexter and others 1982) such inputs are not reflected in these sediment constituents.

The second time-scale is the past decade and one-half as measured in the Mussel Watch monitoring. The basic pattern that emerges is that, with the exception of mercury, lead and zinc, metal concentrations have been stable or, in some cases, cyclic. Statistical testing is required to sort out the significance of these patterns regionally or locally. Mercury and lead (Pb) were presumably declining in the 1980s as a result of control of sources and leaded gasoline, respectively. Sources of zinc could have included automobile tire wear (Christiansen and Guinn 1979). More dramatic were the clearly decreasing trends in concentrations of PAHs and the organochlorine compounds during the 1980s, with most “leveling” off during the 1990s. However, almost all the organic contaminants experienced a five-fold or greater increase at Four Mile Rock during the period 1996-98. This suggests that a significant, presumably local event took place, which disturbed or unleashed a mix of chemicals into northern Elliot Bay. One possibility, for example, is development of the new marina. Since new data is not yet available we don’t know if this was a “spike” or the beginning of a new long-term trend.

Examination and comparison of the relative abundances of individual PAH analytes in the mussels indicates that the low concentrations of T PAHs from mussels at the outer coastal site, Cape Flattery, produce a fingerprint similar to crude oil whereas most of the samples from inside Puget Sound and the Northwest Straits yield patterns more typical of combusted fuels (data not shown here). This is consistent with Lefkovitz and others (1997) who concluded that 80% of the PAH’s in main Basin Cores derived from combustion from 1940 to 1990.

Several metals such as cadmium and arsenic in mussels from the Pacific coast sites may have experienced fluctuations or cycles. Although these need to be examined in more detail, and with a longer time-series, it is possible that these mussels have been recording changing ocean conditions. Cadmium is rich in upwelling seawater (reviewed in Mearns and others 1991) so its variability in mussels from the coast as well as at interior sites (Port Angeles) may be a measure of variability of upwelling and fresh deep water entering the Straits and the Sound. Indeed, using comparable Mussel (ie, oyster) Watch contaminant data from the Gulf of Mexico, Kim and others (1999) found strong relations between oceanic cycles (El Niño, La Niña, etc) and variations in both inorganic contaminants and some pesticides.

Conclusion and Recommendation

It is clear from this cursory review that Puget Sound, and perhaps the Straits, are receiving considerably less chemical contamination than they were 20 to 50 years ago. The dated cores from Puget Sound clearly indicate that most of the materials we consider as pollutants of concern were discharged into the Sound in much greater quantities during the 1940s through 1960s than now (1990s). This means that effective pollution controls were implemented at least as early as the 1960s. Such controls include: cessation of chemical production, sales and/or use (coal and burning—a source of PAHs, 1960's; DDT, 1972; PCBs, 1978; chlordane, 1983 (Shigenaka and others 1990); industrial pre-treatment of raw wastewater (most metals, 1970s and 1980s); source water treatment to reduce erosion of copper from plumbing; closure and/or secondary treatment of pulp mill wastes (late 1960s to mid-1970s; Dexter and others 1984) and secondary treatment of municipal wastewaters (late 1980s to mid-1990s). Most of the reductions apparently occurred well before secondary treatment of municipal wastewater was implemented. During the past two decades, public agencies and industries of coastal Washington have spent billions of dollars to further reduce the input of chemical contaminants to Puget Sound, the Northwest Straits and along the Pacific Coast. The specific extent to which these more recent actions are contributing to reducing contaminant concentrations in biota and sediments of the Sound has been poorly documented in part because monitoring data such as those reported here, has not been vigorously examined and analyzed and also because there have been no recent attempts to document and compare mass contaminant inputs from various sources, such as was done in the early 1980s (Konasewich and others 1982; Dexter and others 1985; Quinlan and others 1985). A renewal of such efforts—comparison of regional chemical monitoring data with estimates of regional contaminant inputs—could point us more directly to actions that have provided benefit and those that have not. It would also be instructive to compare these data with long-term biological data sets such as Nichols (in press).

Perhaps the most important implication of this work is that it provides a basis and stimulus for continued region-wide contaminant monitoring using these or similar tools. There is no assurance that NOAA or any other federal agency will continue to fund such programs into the future. Already Mussel Watch sampling frequency has been reduced to biennial and replication reduced from 3 to 1 composite. This occurred during a period (mid-1990s) when sudden spikes of contamination (PCBs, PAHs, pesticides) reappeared at sites such as Four Mile Rock in Elliot Bay after many years of declining or stable trends of chemical contamination, and after the expenditure of billions of dollars public and industry funds. Presumably such events are indeed spikes and not harbingers of increasing long-term contamination. The only way to find out is to keep monitoring. Such monitoring could go backward as well as forward, making use of even earlier historic data on contaminants in shellfish (Mearns and others 1988). Likewise, it has been over a decade since the last dated cores were taken from Puget Sound (Lefkovitz and others 1997). These cores indicated that at least the bottom of the Sound was heavily contaminated during the 1940s to 1960s and that actions taken during and since that time have reversed the trend. However, that picture stops in 1990. This work supports Yake's (2001) call for more dated core work. New cores, taken every decade or so, could connect recent with past history, provide further measures of progress and warn future generations of a creeping increases of contemporary contaminants.

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